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## GROUND BASED STUDIES OF THERMOCAPILLARY FLOWS IN LEVITATED DROPS

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**ABSTRACT:** Ground-based experiments together with analytical studies are presently being conducted for levitated drops. Both acoustic and electrostatic techniques are being employed to achieve levitation of drops in a gaseous environment. The scientific effort is principally on the thermal and the fluid phenomena associated with the local heating of levitated drops, both at 1-*g* and at low-*g*. In particular, the thermocapillary flow associated with local spot heating is being studied.

Fairly stable acoustic levitation of drops has been achieved with some exceptions when random rotational motion of the drop persists. The flow visualization has been carried out by light scattering from smoke particles for the exterior flow and fluorescent tracer particles in the drop. The results indicate a lack of axial symmetry in the internal flow even though the apparatus and the heating are symmetric.

The theoretical studies for the past year have included fundamental analyses of acoustically levitated spherical drops. The flow associated with a particle near the velocity antinode is being investigated by the singular perturbation technique. As a first step towards understanding the effect of the particle displacement from the antinode, the flow field about the node has been calculated for the first time. The effect of the acoustic field on the interior of a liquid drop has also been investigated. The results predict that the internal flow field is very weak.

### INTRODUCTION

One of the important uses of containerless processing is deep undercooling which can promote certain types of crystal growth and at the same time provide homogeneity of the product. In this regard, there is a strong interest in understanding the basic thermodynamics of such processes as well as the measurement of properties such as thermal conductivity and thermal diffusivity. In this regard, the potential for fundamental advancement in materials processing through containerless experimentation is well recognized.

With ground-based studies, the acoustic and electrostatic levitation offer suitable alternatives to zero- and low-gravity levitation. However, the level of undercooling achievable with the latter is considerably higher but at a much greater cost. Under microgravity conditions, the levitation apparatus is used to stabilize the drops with weak acoustic or electrostatic fields which can afford a lower level of deformation and other disturbances.

### OBJECTIVES

#### EXPERIMENTAL STUDIES

The principal objectives were first to develop the experimental capability to quantitatively characterize thermocapillary flows within freely suspended and spot-heated droplets under the full effect of gravity. The second experimental objective was to identify the direct and indirect effects of the gravitational field on the thermocapillary flow fields internal to the drop. The direct effects are related to the interference by the gravitationally-induced convection, while the indirect effects originate from the additional external convection and the solid body and differential motions induced by the high intensity levitation fields.

## ANALYTICAL STUDIES

The theoretical objectives consist of the development of models that appropriately account for the acoustic field interference and predict the thermocapillary flows for the imposed thermal stimuli. Under the current program for ground-based studies, analytical models of acoustically levitated drops with thermal interaction are being developed with the final aim of having a comprehensive model that would work in conjunction with the experiments. For this purpose, numerical and perturbation solutions for the combined thermal and acoustic fields are being developed.

## SIGNIFICANCE

As stated earlier, containerless processing is useful for deep undercooling which facilitates the production of certain types of crystals with a high degree of uniformity. In particular, there is high potential for the production of technologically useful new materials. The ongoing research will provide fundamental understanding of the Marangoni flows associated with localized heating of levitated drops. In addition, a comprehensive theoretical and experimental system is being developed for the thermodynamic measurement and analysis of materials in containerless environments. For ground based studies where there is interference from the acoustic field, a sound numerical model will provide significant new information about the behavior of these complex systems. Most importantly, the model development along with the experimental studies will represent fundamental groundwork for the measurement of thermophysical properties of undercooled liquids.

## RESEARCH APPROACH

### EXPERIMENTS

The experimental approach is based on Earth-based single-drop levitation using a unique ultrasonic-electrostatic hybrid technique and fluorescent particles suspended in the drop. These tracer particles are illuminated by a laser sheet (or a collimated beam) at the fluorescence excitation wavelength, and the scattered signal is detected at the fluorescence emission wavelength. The particle motion is tracked through CCD cameras and the optically-corrected flow fields are digitally analyzed. The hybrid ultrasonic-electrostatic levitation approach allows the experimental parametric evaluation of the effects of drop distortion and external flows due to acoustic radiation stresses and streaming, and the assessment of the influence of a net electrical charge on the drop surface in the presence of a high static electric field.

### ANALYSIS

The relevant problems can be mathematically described by the Navier-Stokes equations and the energy equation in a coordinate system suitable to the spherical or the spheroidal geometries. With a defined acoustic field and a quantified heat source, approximate analytical methods and numerical methods will be employed.

*Asymptotic Methods:* The perturbation expansions are useful when a small parameter can be identified. Among the various approximations being used include the high frequency acoustic field limit for which various large and small dimensionless parameters have been identified. In particular,  $\varepsilon = U_0/a\omega \ll 1$  and  $M^{-1} \ll 1$ , with  $M^2 = a^2\omega/\nu$ , serve as suitable perturbation parameters. Here,

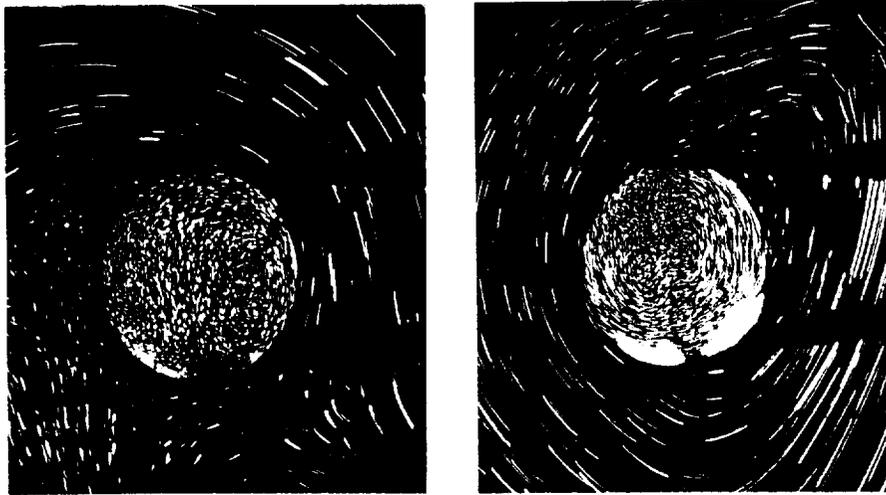


FIGURE 1: Entrained internal flows within droplets suspended in a liquid medium where external streaming flows have been generated.

$U_0$  is the velocity amplitude,  $\omega$  is the frequency,  $\nu$  is the kinematic viscosity and  $a$  is the particle length scale.

*Numerical Methods:* Outside the range of the above approximations, numerical treatment is necessary. In fact, even for the perturbation methods used, some cases of asymptotic matching required numerical procedures. Finite difference techniques would be the most suitable for full numerical treatment of the Navier-Stokes and the energy equations.

## RESULTS

### EXPERIMENTS

*Studies of Acoustic Streaming:* Flow visualization studies of the external forced convective fields generated by the high intensity ultrasound used to levitate millimeter-size droplets under the full effect the Earth gravitational field have been previously reported. The increase in the heat and mass transfer coefficients between a levitated liquid drop and the surrounding gas has been quantitatively evaluated. In addition to affecting the transport process, external flows also induce low velocity internal flows due to entrainment. Arising from the time-varying acoustic motion and from the steady streaming flow component, this entrained flow field is weak for the case of a liquid drop levitated in a gas. It can be substantial, however, for a drop levitated in an immiscible liquid host (see Figure 1). In summary, the assessment is that it will be possible to quantitatively evaluate the influence of external convection on thermocapillary flows in a microgravity environment where the controlled introduction of such external convection can be obtained from a zero base state.

*Internal Flow Visualization in Levitated Drops in 1 G:* An apparatus described in Figure 2 has been used to record the motion of fluorescent tracer particles suspended in the drop liquid. Polystyrene particles of Florida Yellow G from Bangs Laboratories having 0.405 microns in average diameter have been visualized using an Argon Ion (488 nm) laser sheet with variable orientation (the sheet thickness is about 200-300 microns). The scattered light is gathered along two orthogonal views using holographic notch filters to block the elastically scattered light from the drop surface. The liquid used was an aqueous mixture of glycerin and silicone oil (Polydimethylsiloxanes) and

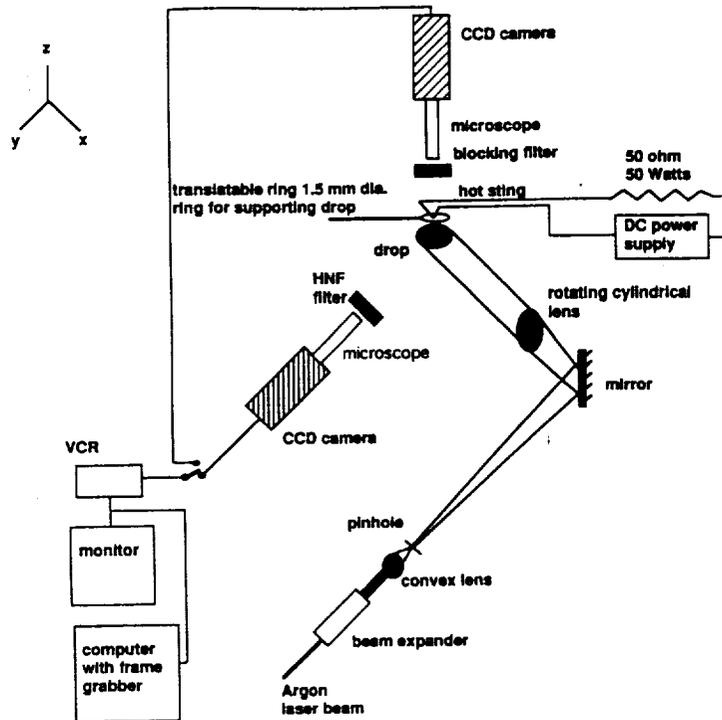


FIGURE 2: Apparatus for flow visualization in a levitated drop.

a focused  $\text{CO}_2$  laser was used to spot heat the levitated drop. The results show that although it was possible to accurately measure the internal flows of isothermal drops, the combination of Earth-based levitation and spot heating induces an uncontrolled torque which drives a random rotational motion of the drops. The digital image processing required in the deconvolution of this rotational motion in order to extract the thermocapillary and buoyancy-driven flows requires substantial computational power, and will be pursued by this experimental effort. Figure 3 shows photographs of time-exposures of the residual motion within a spot-heated drop after heating has stopped. In general, all the recorded flow fields have lacked axial symmetry although both the levitation apparatus and the heating direction are axially symmetrical. Control of drop evaporation has been implemented by maintaining the drop environment at a high humidity, and the Marangoni convection contribution due to evaporation can thus be neglected. Ongoing and future studies will include the measurement of flows within drastically flattened drops to constrain the flows in a two dimensional plane, the implementation of total electrostatic levitation of charged droplets, and an automated digital data reduction and analysis.

*Glovebox Low-Gravity Demonstration:* A Glovebox flight investigation tentatively scheduled for April 1997 has been initiated to assess the capability for ultrasonic positioning in microgravity for drop internal flow measurement. A compact ultrasonic positioner has been designed and integrated with laser diode illumination in order to experimentally demonstrate the rotation control of freely suspended drops in low gravity and to obtain preliminary flow field measurements for sting-heated droplets. A laboratory breadboard is already available, and the flight unit is currently nearing design completion.

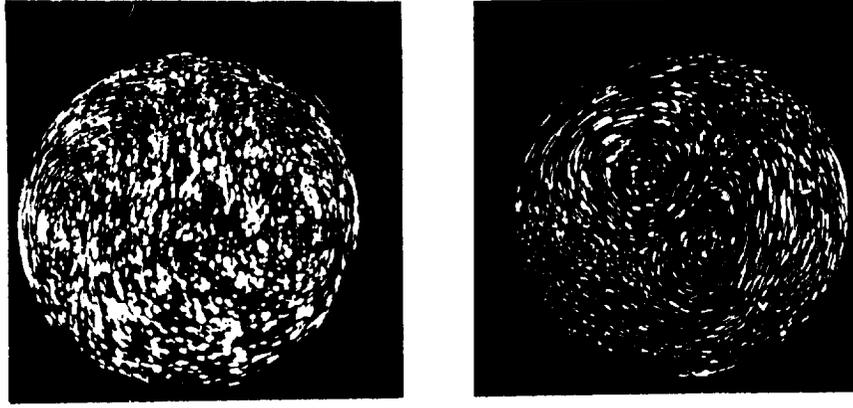


FIGURE 3: Time exposure (10 sec) photographs of slow internal flows in levitated drops containing tracer particles. These are residual flows observed after the spot heating has been terminated.

#### ANALYSIS

The theoretical analysis of the flow field associated with a levitated spherical particle requires a detailed study of the fluid mechanics of streaming flows. In particular, the disturbance due to the presence of the particle needs to be investigated. Riley [2] has given a singular perturbation solution which may be used for the flow associated with a solid particle at the velocity antinode. Presently, this has been extended to the case of a fluid particle. In addition, the technique has been used to calculate the flow for a solid particle at the velocity node.

*Spherical Particle in an Acoustic Field:* As mentioned before, Riley [2] gave the solution for the flow field associated with a sphere oscillating in a fluid. This solution is applicable to a particle at the velocity antinode of a standing wave. Attempts are being made to derive a rigorous solution for the flow with a particle displaced from the antinode. For a particle of radius  $a$  and a displacement  $Z_0$ , with the limiting approximation  $ka \ll 1$ , the 'far-field' velocity field is given by

$$u = A \cos [k(z + Z_0)] e^{i\tau} = A [\cos kZ_0 + (kz) \sin kZ_0 + \dots], \quad (1)$$

$$\psi = (A \cos kZ_0) \psi^{(0)} + (Ak \sin kZ_0) \psi^{(1)} + \text{nonlinear terms} \quad (2)$$

where  $R = U_\infty a / \nu$ ,  $M = i\omega a^2 / \nu$ ,  $\psi^{(0)}$  is the stream function for the case when the particle is at the velocity antinode and  $\psi^{(1)}$  for the antinode. The expression for  $\psi^{(0)}$  is already available for the limit  $\varepsilon = R|M|^{-2} \ll 1$ . The new results include the perturbation solution for  $\psi^{(1)}$  which has been calculated to the second order. However, only the first order results are presented here.

Outer Solution:

$$\begin{aligned} \psi^{(1)} \rightarrow \phi^{(1)} &= \frac{1}{2}(r^3 - r^{-2})\bar{\mu}(1 - \bar{\mu}^2) \cos \tau \\ &+ \varepsilon r^{-4}\bar{\mu}(1 - \bar{\mu}^2) \left[ -\frac{5}{2}(R_s^{-\frac{1}{2}} r^2 \cos(\tau - \frac{1}{4}\pi) + \frac{75}{112}(r^2 - 1) \left\{ r^2 + \frac{3}{2}(7\bar{\mu}^2 - 3) \right\} \right] + \dots \end{aligned} \quad (3)$$

Within the Shear-Wave Layer:

$$\begin{aligned} \psi^{(1)} \rightarrow \Phi^{(1)} &= \bar{\mu}(1 - \bar{\mu}^2) \left\{ \frac{5}{2} \left[ \eta - \frac{1}{2}(1 - i) \left( e^{-(1+i)\eta} \right) \right] \cos \tau \right. \\ &+ \varepsilon \left[ \left( \frac{325}{32} - \frac{75}{16}\eta \right) - \frac{25}{16} \cos(2\tau + \frac{1}{4}\pi) \right. \\ &\left. \left. + \bar{\mu}^2 \left( -\frac{875}{32} + \frac{225}{16}\eta + \frac{275\sqrt{2}-225}{32\sqrt{2}} \cos(2\tau + \frac{1}{4}\pi) \right) \right] \right\} + \dots, \end{aligned} \quad (4)$$

where  $\varepsilon = R|M|^{-2}$ ,  $R_s = \varepsilon R$ ,  $\bar{\mu} = \cos \theta$ , and  $\eta = \frac{1}{2}(r-1)|M|$ .

*Internal Circulation in a Drop in an Acoustic Field:* The investigation of the internal flow in a drop at the antinode of a standing wave has been carried out by singular perturbation approach of Riley [2]. At the fluid-fluid interface, the stress and velocity continuity condition are applied. The results for the drop phase stream function to the leading order are as follows:

$$\hat{\psi}^{(0)} = c^* \left[ r^2 \left( \frac{1}{\widehat{M}} + 1 \right) e^{-\widehat{M}} - r^2 \left( \frac{1}{\widehat{M}} - 1 \right) e^{\widehat{M}} \left( \frac{1}{\widehat{M}r} + 1 \right) e^{-\widehat{M}r} + \left( \frac{1}{\widehat{M}r} - 1 \right) e^{\widehat{M}r} \right] (1 - \bar{\mu}^2) e^{i\tau}, \quad (5)$$

where

$$c^* \simeq \frac{\frac{3}{2} \left( 1 - \frac{\widehat{M}}{\widehat{M} + \lambda M} \right)}{-\widehat{M} e^{\widehat{M}}}, \quad \widehat{M}^2 = \frac{i\omega a^2}{\bar{\nu}},$$

$\bar{\nu}$  is the kinematic viscosity of the liquid phase, and  $\lambda = \mu/\bar{\mu}$  is the ratio of the gas to the liquid dynamic viscosities. It is observed that the strength of this internal flow behaves like  $\sim e^{-\widehat{M}/\widehat{M}}$ . For large  $\widehat{M}$ , this is very weak. The reason for such weak flow is mainly the recirculating Stokes layer which is very thin. Since there are opposing velocities within it, a very large shear stress is required to sustain its motion. The system cannot afford such a large shear stress at the interface and the result is weak internal circulation. The large drop viscosity as compared with the gas also has a role in weakening this circulation.

## CONCLUSION

The analytical and experimental studies of drops in an acoustic field have led to several new results. These are briefly summarized here.

1. The theoretical analysis of the flow field inside an acoustically levitated spherical drop shows that the internal circulation is quite weak. This is owing to the thin recirculating shear layer which cannot sustain the large stresses that would arise if there were substantial internal flow. While experimental studies also show a weak internal circulation, the magnitude has not yet been verified to be in agreement with the extremely low predictions. However, a liquid drop in a liquid host is seen to experience substantial internal circulation.

The external flow field associated with a particle between the velocity antinode and the node of a standing wave can be described as a combination of the independent fields about these point. In addition, there are some nonlinear interaction terms that are of the same order.

2. It has been possible to accurately measure the internal flows of isothermal drops. The results of the combination of the Earth-based levitation and the spot heating induces an uncontrolled torque that drives a random solid-body like rotational motion of the drops.

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